

## REMARKS

Claims 1-5, 7, 9-13, 15, 17-18 and 20 remain pending in the application. Claims 6, 8, 14, 16 and 19 were cancelled in a prior amendment. No claims are amended in the present reply. The Examiner is requested to reconsider and withdraw the rejections in view of the remarks contained herein.

### 1. REQUEST FOR RECONSIDERATION OF FINALITY

Applicants request withdrawal of finality of the latest Office Action, dated June 15, 2009. Applicants submit that finality is premature in the present case as the amendments provided in Applicants' response of March 11, 2009 were clarifying amendments to address issues under 35 U.S.C. § 112. These amendments were not made to traverse prior art, did not change the scope of the claims, and the new search and present application of new art rejections were not necessitated by the amendments. For instance, a new rejection based on the Pettit reference is applied against claims 4 and 5, which (along with claim 1) were amended to clarify the identity of particular enclosures and vents, as illustrated in the Amendment filed March 11, 2009. Traversals of the art-based rejections in the response were based on unamended claim features. Hence Applicants submit the final status of the latest Office Action is premature.

### 2. REJECTIONS UNDER 35 U.S.C. § 112

Claims 9 & 20 stand rejected under 35 U.S.C. § 112, second paragraph, as allegedly indefinite for failing to particularly point and distinctly claim the subject matter which Applicant regards as the invention. This rejection is respectfully traversed.

Claims 9 and 20 include “a third enclosure that encompasses at least one of the first enclosure and the second enclosure.” The quoted passage is clear and definite. The plain language of this phrase taken in view of the respective base claims and the specification indicates that the third enclosure encompasses the first enclosure *or* encompasses the second enclosure *or* encompasses both the first and second enclosures. The expression “at least one of A and B” is common claim parlance that refers to definite alternatives A, B, or both A and B without using the conjunction “or,” as illustrated in the preceding sentence. As a matter of plain language, “at least one of” a group of items means one or more.

Applicants request reconsideration of the claims and withdrawal of the rejection.

### **3. REJECTION UNDER 35 U.S.C. § 102 / 103 – PETTIT**

Claims 1, 2, 4, 5, 7, 11-13 & 15 stand rejected under 35 U.S.C. § 102(e) as allegedly anticipated by or, in the alternative, under 35 U.S.C. 103(a) as allegedly obvious over Pettit (U.S. Pub. No. 2005/0058861). This rejection is respectfully traversed.

Independent claims 1 and 11 are not anticipated or obvious over Pettit as the reference fails to provide for a fuel cell with first and second passive vents, and further fails to disclose such a vent configured to maintain hydrogen below about 4 percent. Claim 1 is drawn to a fuel cell that includes “a first passive hydrogen vent configured to vent hydrogen from the first enclosure without reliance upon any electrical device and configured to maintain the hydrogen concentration within the first enclosure below about 4 percent” and “a second hydrogen vent configured to vent hydrogen from the second enclosure.” Claim 11 is drawn to a method of manufacturing a fuel cell that includes “providing a first passive hydrogen vent in the first enclosure, the first passive hydrogen vent configured to passively maintain the level of hydrogen

which leaks into the first enclosure below a concentration level of about 4 percent" and "providing a second hydrogen vent in the second enclosure." These features are absent from the Pettit disclosure.

Anticipation requires each and every element as set forth in the claim to be expressly or inherently described in a single prior art reference. *Verdegaal Bros. v. Union Oil Co. of California*, 814 F.2d 628, 631, 2 USPQ2d 1051, 1053 (Fed. Cir. 1987). Since the passive vent features are missing from Pettit, the present claims are consequently novel.

The present claims include a passive hydrogen vent, as illustrated on page 8, lines 3-20 of the present specification:

The hydrogen vent 88 of this embodiment is adapted to allow hydrogen to pass therethrough while simultaneously preventing any coolant (including evaporating coolant vapor) from passing therethrough. In addition, the hydrogen vent 88 is preferably adapted to prevent a flame front from passing into the enclosure through the hydrogen vent 88. The hydrogen vent 88 provides pores (represented by the cross-hatching) which are sufficiently large to allow hydrogen molecules to pass therethrough. The pores are also preferably sufficiently small that coolant, including coolant vapors, cannot pass therethrough. Thus, the hydrogen vent 88 is passive. As used herein "passive" means that the hydrogen vent does not require any electrical or other active components to function. For example, the hydrogen vent 88 requires no electrical components such as a sensor, controller, or fan are required.

In addition, the hydrogen vent 88 is preferably adapted to passively vent hydrogen such that the hydrogen remains below about 4 percent within the enclosure 72; and more preferably, below about 1 percent. The hydrogen vent 88 is preferably made of a porous material selected from the group consisting of cellulose, plastic (for example, a foamed plastic) or metal (for example, a sintered metal).

This is in contrast to "[p]revious approaches to venting accumulated hydrogen from a fuel cell [that] have included use of a hydrogen detector and/or fan to actively ventilate the reservoir. These electrical devices consume electricity, reducing the efficiency of the fuel cell. In addition, they can result in ventilating coolant vapor, thereby evaporating the coolant." Specification page

3, lines 4-8. Pettit provides an example of these “previous approaches” to venting hydrogen that use electrical components such as a sensor (e.g., hydrogen detector), controller, or fan to provide a ventilation stream. Obviously, in view of this discussion, the active vents must be considered to include more than an opening, as the detectors, fans, etc. are not the opening itself. The Examiner’s interpretation offered on pages 2-3 of the office action quotes a relevant passage from the specification, but then promptly misconstrues it. If such electrical components as are mentioned in the passage operate in conjunction to vent hydrogen, then the vent is an active one. The Examiner’s interpretation gives “passive vent” the same meaning as “active vent;” so such a definition is unreasonable and specious, and it cannot hold. The Pettit reference therefore does not provide for the presently claimed passive hydrogen vent, and as further illustrated below, the Pettit reference does not actually vent hydrogen (hydrogen is instead combusted), and providing a passive vent in the Pettit apparatus would further serve to contravene operation of the Pettit apparatus.

First, Pettit does not provide a passive vent as alleged by the rejection. Pettit discloses a fuel cell stack enclosure to contain migrating hydrogen and to direct the migrating hydrogen to a desired location or component. Pettit paragraph [0005]. A hydrogen sensor located in the enclosure is used to detect the presence of hydrogen so that a corrective action can be commenced. Pettit paragraph [0005]; and abstract. A ventilation stream flows through the enclosure and exits the enclosure through the outlet. Pettit paragraph [0006]. The ventilation stream is induced by operation of the compressor of the oxidant delivery system, where the ventilation stream is directed through an outlet in the enclosure that contains a hydrogen sensor, operable to both detect the presence of hydrogen and to consume the hydrogen within the ventilation stream prior to being exhausted from the enclosure. Pettit paragraph [0047]. The

ventilation stream can also be induced by operation of a fan driven by a motor which operates independently of the operation of the oxidant delivery system. Pettit paragraph [0047]. Consequently, the Pettit reference does not disclose a passive vent as found in the present claims.

Second, the exhaust stream 70 of Pettit cannot function as an alleged “second hydrogen vent.” The exhaust stream 70 comes from combustor system 46 which removes the hydrogen provided by anode effluent stream 32. Pettit paragraph [0031]: and see FIG. 1. The “combustor system 46 [is] operable to consume the remaining hydrogen in anode effluent stream 32.” Pettit paragraph [0021]. Thus, no hydrogen is vented via the exhaust stream 70 and accordingly it is not a “second hydrogen vent.”

Third, Pettit combusts all hydrogen collected via the ventilation stream so that no hydrogen is actually vented from the fuel cell system. As best can be ascertained from the rejection, the vent the Examiner identifies in Pettit includes the ventilation flows 41 accumulating and passing through the catalytic combustion element 72 and exiting the outlet 42 of enclosure 40. Pettit FIG. 1. As described in Pettit paragraph [0031]:

The catalytic combustion of the hydrogen within ventilation stream 41 on catalytic combustion element 72 consumes the hydrogen in ventilation stream 41 prior to ventilation stream 41 exiting enclosure 40. The catalytic combustion of hydrogen also increases the temperature of catalytic combustion element 72 and of ventilation stream 41 that is detected by temperature sensing device 74 which then sends a signal to the microprocessor indicating the presence of hydrogen within ventilation stream 41 and the combustion of the hydrogen. Thus, hydrogen sensor 44 is operable to simultaneously detect the presence of and consume hydrogen within ventilation stream 41. The ventilation stream 41 then joins anode effluent stream 32 to form combustion stream 68 that flows through combustor system 46 for removal of the hydrogen (provided by anode effluent stream 32) and is exhausted as exhaust stream 70.

As such, this vent is not passive, as provided by the present claims, and what is more, no hydrogen is actually vented. The hydrogen is detected and consumed prior exiting the outlet 42 of enclosure 40. Pettit FIG. 1. The reference therefore cannot anticipate the present claims.

Finally, because Pettit does not disclose any passive vent, it also does not disclose the further refinement of a passive vent that maintains the concentration of hydrogen below 4 percent.

Turning now to the obviousness rejection, to establish a *prima facie* case of obviousness, the cited prior art must teach or suggest all of the claim limitations. *In re Vaeck*, 947 F.2d 488, 20 USPQ2d 1438 (Fed. Cir. 1991). When a reference is missing claimed features, obviousness requires an apparent reason based either in the reference or the general knowledge in the art by which a skilled artisan would modify the reference to include the missing subject matter. See *In re Vaeck*, 947 F.2d 488, 20 USPQ2d 1438 (Fed. Cir. 1991); and *KSR Int'l Co. v. Teleflex Inc.*, 550 U.S. 398, 82 USPQ2d 1385, 1396 (2007) (obviousness includes determining whether there was an apparent reason to combine the known elements in the fashion claimed). The apparent reason to combine or modify the reference should be made explicit in order to facilitate review. *KSR Int'l Co. v. Teleflex Inc.*, 550 U.S. 398, 82 USPQ2d at 1396; *In re Kahn*, 441 F3d 977, 988, 78 USPQ2d 1329, 1336 (Fed. Cir. 2006) (“[R]ejections on obviousness grounds cannot be sustained by mere conclusory statements; instead, there must be some articulated reasoning to support the legal conclusion of obviousness.”). In the present case, there is no way a skilled artisan could modify Pettit to recreate the present claims without contravening the teachings and operation of the Pettit disclosure.

Modification or adaptation of the Pettit apparatus to include a passive hydrogen vent would contravene the Pettit disclosure and likely render aspects of the apparatus inoperable.

Replacement of the outlet including the catalytic combustion element 72, combustor 46, and associated features including the anode effluent stream 32 (which contains hydrogen to be combusted) of Pettit with a passive hydrogen vent would defeat the Pettit teachings by not combusting the collected hydrogen; i.e., Pettit burns the hydrogen, there is no reason to passively release it. Moreover, use of a passive hydrogen vent would require removal or modification of the compressor, controller, and/or fan responsible for providing the ventilation streams 41. Such modifications are contrary to the function and goals of the Pettit disclosure and are counter to a case of obviousness. See *In re Ratti*, 270 F.2d 810, 123 U.S.P.Q. 349 (CCPA 1959) (if the proposed modification or combination of the prior art would change the principle of operation of the prior art invention being modified, then the teachings of the references are not sufficient to render the claims obvious).

The present claims are therefore novel and unobvious over the Pettit reference. Accordingly, Applicants request reconsideration of the claims and withdrawal of the rejection.

#### **4. REJECTION UNDER 35 U.S.C. § 103 – PETTIT & BUZZELLI**

Claims 10 & 17 stand rejected under 35 U.S.C. § 103(a) as allegedly unpatentable over Pettit (U.S. Pub. No. 2005/0058861) in view of Buzzelli (U.S. Pat. No. 4,168,349). This rejection is respectfully traversed.

The shortcomings of the Pettit disclosure are detailed in Section 3. Buzzelli is provided for teaching an iron/air battery cell having a sintered ceramic vent that acts as a flame and explosion barrier. Buzzelli col. 2, lines 55-60. However, the Buzzelli reference fails to provide the missing features, teachings, and reasons necessary to overcome the deficiencies of Pettit. Namely, the reference combination does not provide for the presently claimed passive hydrogen

vent, does not actually vent hydrogen (hydrogen is instead combusted), and providing a passive vent in the Pettit would further serve to contravene operation of the Pettit apparatus, an action the references provide no reason to take . Claims 10 and 17 are therefore patentable over the combination of Pettit and Buzzelli.

Accordingly, Applicants request reconsideration of the claims and withdrawal of the rejection.

## **5. REJECTION UNDER 35 U.S.C. § 103 – PETTIT & GENC**

Claim 3 stand rejected under 35 U.S.C. § 103(a) as allegedly unpatentable over Pettit (U.S. Pub. No. 2005/0058861) in view of Genc (U.S. Pub. No. 2002/0160245). This rejection is respectfully traversed.

The shortcomings of the Pettit disclosure are illustrated in Section 3. Genc is provided for disclosing a release valve 30 having a membrane 40 operable to permit passage of gas and inhibit passage of liquid at a first pressure. Genc paragraphs [0019]-[0020] on pages 1-2; and Figure 2. The membrane may be constructed of various polymers. Genc paragraph [0021] on page 2. However, the Genc reference fails to provide the missing features, teachings, and reasons necessary to overcome the deficiencies of Pettit, as illustrated in Section 2. Namely, the reference combination does not provide for the presently claimed passive hydrogen vent, does not actually vent hydrogen (hydrogen is instead combusted), and providing a passive vent in the Pettit apparatus would further serve to contravene operation of the Pettit apparatus. Claim 3 is therefore patentable over the combination of Pettit and Genc.

Accordingly, Applicants request reconsideration of the claims and withdrawal of the rejection.

## **6. REJECTION UNDER 35 U.S.C. § 103 – PETTIT, BUZZELLI & GENC**

Claim 18 stand rejected under 35 U.S.C. § 103(a) as allegedly unpatentable over Pettit (U.S. Pub. No. 2005/0058861) of view of Buzzelli (U.S. Pat. No. 4,168,349) as applied to claim 17 above and further in view of Genc (U.S. Pub. No. 2002/0160245). This rejection is respectfully traversed.

The shortcomings of the Pettit disclosure are illustrated in Section 3, the Buzzelli disclosure is illustrated in Section 4, the Genc disclosure is illustrated in Section 5. However, the Buzzelli and Genc references fail to provide the missing features, teachings, and reasons necessary to overcome the deficiencies of Pettit, as illustrated in Section 3. Namely, the reference combination does not provide for the presently claimed passive hydrogen vent, does not actually vent hydrogen (hydrogen is instead combusted), and providing a passive vent in the Pettit apparatus would further serve to contravene operation of the Pettit apparatus. Claim 18 is therefore patentable over the combination of Pettit, Buzzelli, and Genc.

Accordingly, Applicants request reconsideration of the claims and withdrawal of the rejection.

## **7. REJECTION UNDER 35 U.S.C. § 103 – PETTIT & WALSH**

Claims 9 & 20 stand rejected under 35 U.S.C. § 103(a) as allegedly unpatentable over Pettit (U.S. Pub. No. 2005/0058861) in view of Walsh (U.S. Pub. No. 2003/0118881). This rejection is respectfully traversed.

The shortcomings of the Pettit disclosure are illustrated in Section 3. Walsh is provided for disclosing that “an integrated fuel cell system may or may not be housed in a common

enclosure.” Walsh paragraph [0009]; and see discussion of Walsh in Section 8 below. However, the Walsh reference fails to provide the missing features, teachings, and reasons necessary to overcome the deficiencies of Pettit, as illustrated in Section 3. Namely, the reference combination does not provide for the presently claimed passive hydrogen vent, does not actually vent hydrogen (hydrogen is instead combusted), and providing a passive vent in the Pettit apparatus would further serve to contravene operation of the Pettit apparatus. Claims 9 and 20 are therefore patentable over the combination of Pettit and Walsh.

Accordingly, Applicants request reconsideration of the claims and withdrawal of the rejection.

## **8. REJECTION UNDER 35 U.S.C. § 103 – WALSH**

Claims 1, 2, 4, 7, 9, 11, 12, 16 & 20 stand rejected under 35 U.S.C. § 103(a) as allegedly unpatentable over Walsh (U.S. Pub. No. 2003/0118881). This rejection is respectfully traversed.

Independent claims 1 and 11 are not obvious over Walsh as the reference fails to provide for any of the passive hydrogen vent limitations, the hydrogen concentration limitations, the first and second hydrogen vents, and the first and second enclosures. Moreover, there is no apparent reason for a skilled artisan to include the missing subject matter in the context of Walsh. Claim 1 is drawn to a fuel cell that includes “a first passive hydrogen vent configured to vent hydrogen from the first enclosure without reliance upon any electrical device and configured to maintain the hydrogen concentration within the first enclosure below about 4 percent” and “a second hydrogen vent configured to vent hydrogen from the second enclosure.” Claim 11 is drawn to a method of manufacturing a fuel cell that includes “providing a first passive hydrogen vent in the first enclosure, the first passive hydrogen vent configured to passively maintain the level of

hydrogen which leaks into the first enclosure below a concentration level of about 4 percent" and "providing a second hydrogen vent in the second enclosure." These features are absent from the Walsh disclosure.

Walsh cannot establish a *prima facie* case of obviousness as the reference is missing the aforementioned claimed features. Walsh is provided for disclosing a vent 114 in a water tank 102. Walsh page 3, paragraph [0032]; and Figures 1-3. The Walsh disclosure has several shortcomings and flaws.

First, the reference provides that condensate in the anode feed 342 and the anode exhaust 344, which may contain combustible gases (i.e., hydrogen), may be dealt with in two different ways. One way is to include water traps, such as 348 and 352, on drain lines to send water to the water tank 302 and thereby limit exposure of the water tank to such hydrogen-containing streams. Walsh page 4, paragraph [0037]; and Figure 3. The other way is by continually purging the atmosphere of the water tank with an air stream to prevent buildup of combustible gases, obviating the need for the water traps. Walsh page 4, paragraph [0037]. Continually purging the atmosphere of the water tank with an air stream is actively forcing gas out through the vent, meaning the Walsh vent is not a passive vent. Cf. present specification at page 8, 11-14: "As used herein 'passive' means that the hydrogen vent does not require any electrical or other active components to function. For example, the hydrogen vent 88 requires no electrical components such as a sensor, controller, or fan are required." The Examiner's interpretation gives "passive vent" the same meaning as "active vent," which is incorrect in view of these disclosures and as illustrated in Section 3, above.

Second, Walsh's water tank is not holding coolant, it is holding condensate. The water tank 102 is not a coolant reservoir and is not even part of a coolant flow path. Instead, the Walsh

teachings are directed toward removing condensate from a process stream in a fuel cell system. Walsh page 1, paragraph [0008]. For example, condensate can interfere with flow of process streams and can build to levels that can flood portions of the system, and can cause problems if allowed to freeze. Walsh page 1, paragraph [0008].

Third, the Walsh water tank has the cathode gas stream ( $O_2$ ) fed into it, not the anode gas stream ( $H_2$ ). Walsh page 3, paragraph [0032]; and see Figures 1-3. For example, the cathode inlet conduit 104 and outlet gas conduit 106 are connected to the water tank 102; note also that the water tank 102 is separate from the fuel cell stack 100. The vent 114 allows gas to exit as it circulates through the tank 102. Thus, the cathode streams provided to the water tank 102 in Walsh do not even contain hydrogen; i.e., there is no hydrogen to vent.

Fourth, as stated by the present rejection, “Walsh is silent to using the collected water in the reservoir as a coolant.”

Nonetheless, the rejection posits that “it is well-known in the art to collect bi-product [sic] water and condensate water from the fuel cell and exhaust streams and use the water as part of a cooling system.” Office Action dated June 15, 2009 page 7, lines 17-18. At the outset, Applicants submit that the Examiner’s conjecture is both unsupported and is in fact erroneous. Applicants request that the Examiner submit an affidavit supporting his personal knowledge that it is well-known in the art to collect byproduct condensate water and use such water in a cooling system for a fuel cell, as required under 37 C.F.R. § 1.104 (d)(2), or otherwise support that conjecture with evidence.

On the other hand, if the Examiner is taking official notice of the above quoted subject matter, Applicants submit that such official notice is improper in the present case as official notice cannot be taken of the state of the art or where the alleged “common knowledge” forms

part of the principal evidence upon which a rejection is based. See *In re Eynde*, 480 F.2d 1364, 1370, 178 USPQ 470, 474 (CCPA 1973) (“[W]e reject the notion that judicial or administrative notice may be taken of the state of the art. The facts constituting the state of the art are normally subject to the possibility of rational disagreement among reasonable men and are not amenable to the taking of such notice.”). In particular, it is never appropriate to rely solely on “common knowledge” in the art without evidentiary support in the record, as the principal evidence upon which a rejection is based. See *In re Zurko*, 258 F.3d 1379, 1385, 59 USPQ2d 1693, 1697 (Fed. Cir. 2001) (“[T]he Board cannot simply reach conclusions based on its own understanding or experience—or on its assessment of what would be basic knowledge or common sense. Rather, the Board must point to some concrete evidence in the record in support of these findings.”).

Applicants further submit that the proposed use of the water condensate in a coolant system is contrary to the Walsh disclosure and the general art of coolants and fuel cell systems. As evidence, Applicants direct the Examiner’s attention to Walsh paragraph [0008] where the reference recognizes that water “condensate can . . . cause problems if allowed to freeze (e.g., in an outdoor unit that is not in service).” This is one reason why Walsh collects and removes the water condensate. See also, for example, U.S. Patent No. 7,393,464, describing an alkylene glycol coolant used in a fuel cell; a hardcopy is enclosed with the present reply. One would not want to dilute a coolant mixture by adding collected condensate water as such dilution would reduce the effectiveness of freezing point depressants and additives.

The rejection also fails to provide any guidance, instruction, or analysis as to how a skilled artisan would adapt the Walsh disclosure to accommodate a cooling system and perform the requisite engineering and plumbing in of the byproduct water and condensate water to such a cooling system. Such details are necessary for a case of obviousness. See *In re Kahn*, 441 F.3d

977, 988, 78 USPQ2d 1329, 1336 (Fed. Cir. 2006) (“[R]ejections on obviousness grounds cannot be sustained by mere conclusory statements: instead, there must be some articulated reasoning to support the legal conclusion of obviousness.”). MPEP § 2143 states that “[t]he key to supporting any rejection under 35 U.S.C. 103 is the clear articulation of the reason(s) why the claimed invention would have been obvious,” which should be made explicit, as directed by *KSR Int'l Co. v. Teleflex Inc.* This burden is not met in the present rejection based on Walsh and the allegation that it is “well-known in the art to collect bi-product [sic] water and condensate water from the fuel cell and exhaust streams and use the water as part of a cooling system.” Office Action dated June 15, 2009 page 7, lines 17-18.

Moreover, as noted in MPEP 2143.01(IV), a statement that modifications of the prior art to meet the claimed invention would have been “well within the ordinary skill of the art at the time the claimed invention was made” because the references relied upon teach that all aspects of the claimed invention were individually known in the art is not sufficient to establish a *prima facie* case of obviousness without some objective reason to combine the teachings of the references. *Ex parte Levengood*, 28 USPQ2d 1300 (BPAI 1993).

Accordingly, Walsh cannot support a case of obviousness and Applicants request reconsideration of the claims and withdrawal of the rejection.

## **9. REJECTION UNDER 35 U.S.C. § 103 – WALSH & GRASSO**

Claims 1, 2, 4, 7, 9, 11, 12, 16 & 20 stand rejected under 35 U.S.C. § 103(a) as allegedly unpatentable over Walsh (U.S. Pub. No. 2003/0118881) in view of Grasso (U.S. Pub. No. 2003/023228). This rejection is respectfully traversed.

The shortcomings of the Walsh disclosure are illustrated above in Section 8. Grasso discloses a liquid-gas separator assembly for separating gas bubbles from a liquid coolant. Grasso abstract. Separated gas is pulled through a line 26 by vacuum pump 25 to an accumulator 20 and vented through line 21. Grasso paragraph [0014]. In contradistinction to the rejection, Grasso is silent with respect to running the process continuously and there is no mention of when or how often the accumulator 20 is purged to vent gas. There is no appreciation of accumulating hydrogen gas and there is no indication that operation of the Grasso assembly would in fact mitigate hydrogen build-up to levels of 4% or 1%. It may be just as possible for the accumulator 20 to fill with separated gas, such as hydrogen, prior to any venting, as the Grasso disclosure is simply silent with respect to hydrogen.

Grasso also fails to supply the missing subject matter and cure the deficiencies of Walsh as previously illustrated. Namely, the reference combination lacks a passive vent, Walsh's water tank is not holding coolant – it is holding condensate, the Walsh water tank has the cathode gas stream ( $O_2$ ) fed into it, not the anode gas stream ( $H_2$ ), and the reference combination is silent with respect to using the collected water in the reservoir as a coolant.

In addition, there is no disclosure in Walsh or Grasso as to how or why a skilled artisan would combine the condensate water collection system of Walsh (which makes no mention of coolant) with the liquid-gas separator assembly (coolant mixture separator) from Grasso. The rejection fails to explicitly provide any way a skilled artisan would combine the separate disclosures, and there no illustration of how and where the two systems could be connected and plumbed together to effectively operate. The instant claims are therefore patentable over the combination of Walsh and Grasso.

Accordingly, Applicants request reconsideration of the claims and withdrawal of the rejection.

#### **10. REJECTION UNDER 35 U.S.C. § 103 – WALSH & NODA**

Claims 1, 2, 4, 7, 9, 11, 12, 16 & 20 stand rejected under 35 U.S.C. § 103(a) as allegedly unpatentable over Walsh (U.S. Pub. No. 2003/0118881) in view of Noda (U.S. Pat. No. 5,623,390). This rejection is respectfully traversed.

The shortcomings of the Walsh disclosure are illustrated in Section 8. Noda is provided for teaching that hydrogen gas should be vented from a computer having a nickel-hydrogen battery so that it does not reach the explosive concentration of 4%. Noda col. 9, lines 7-27. Even so, Noda does not relate this problem to fuel cells, nor does Noda solve a problem of accumulation of H<sub>2</sub> in a coolant system or coolant reservoir, nor does Noda provide any guidance regarding the configuration of vents and enclosures within fuel cells and cooling systems. Catastrophic battery failure is an entirely different problem than the issues addressed by the present claims.

Noda fails to address the several shortcomings of Walsh and provides no means or suggestion to cure Walsh's deficiencies. Namely, the reference combination lacks a passive vent, Walsh's water tank is not holding coolant – it is holding condensate, the Walsh water tank has the cathode gas stream (O<sub>2</sub>) fed into it, not the anode gas stream (H<sub>2</sub>), and the reference combination is silent with respect to using the collected water in the reservoir as a coolant.

As such, the present claims are not obvious over the combined references. Applicants request reconsideration of the claims and withdrawal of the rejection.

## **11. REJECTION UNDER 35 U.S.C. § 103 – WALSH, GRASSO, & NODA**

Claims 1, 2, 4, 7, 9, 11, 12, 16 & 20 stand rejected under 35 U.S.C. § 103(a) as allegedly unpatentable over Walsh (U.S. Pub. No. 2003/0118881) and Grasso (U.S. Pub. No. 2003/023228) in view of (U.S. Pat. No. 5,623,390). This rejection is respectfully traversed.

The shortcomings of the Walsh disclosure are illustrated in Section 8, the Grasso disclosure in Section 8, and the Noda disclosure in Section 10. Addition of the Grasso and Noda disclosures fails to supply the missing subject matter of Walsh and does not provide the means to cure the Walsh deficiencies. Namely, the reference combination lacks a passive vent. Walsh's water tank is not holding coolant – it is holding condensate, the Walsh water tank has the cathode gas stream ( $O_2$ ) fed into it, not the anode gas stream ( $H_2$ ), and the reference combination is silent with respect to using the collected water in the reservoir as a coolant.

As such, the present claims are not obvious over the combined references. Applicants request reconsideration of the claims and withdrawal of the rejection.

## **12. REJECTION UNDER 35 U.S.C. § 103 – WALSH, NODA & BUZZELLI**

Claims 10 & 17 stand rejected under 35 U.S.C. § 103(a) as allegedly unpatentable over Walsh (U.S. Pub. No. 2003/0118881) in view Noda (U.S. Pat. No. 5,623,390) as applied to claims 1 & 16 respectively and further in view of Buzzelli (U.S. Pat. No. 4,168,349). This rejection is respectfully traversed.

The shortcomings of the Walsh disclosure are illustrated in Section 8, the Noda disclosure in Section 10, and the Buzzelli disclosure in Section 4. Combination of the collected teachings fails to provide for a case of obviousness as Noda and Buzzelli do not account for the flaws of the primary reference of Walsh. Namely, the reference combination lacks a passive

vent, Walsh's water tank is not holding coolant – it is holding condensate, the Walsh water tank has the cathode gas stream ( $O_2$ ) fed into it, not the anode gas stream ( $H_2$ ), and the reference combination is silent with respect to using the collected water in the reservoir as a coolant.

As such, the present claims are not obvious over the combined references. Applicants request reconsideration of the claims and withdrawal of the rejection.

### **13. REJECTION UNDER 35 U.S.C. § 103 – WALSH, NODA & GENC**

Claims 3, 5 & 13-15 stand rejected under 35 U.S.C. § 103(a) as allegedly unpatentable over Walsh (U.S. Pub. No. 2003/0118881) in view Noda (U.S. Pat. No. 5,623,390) as applied to claims 1, 2, 4 & 12 respectively and further in view of Genc (U.S. Pub. No. 2002/0160245). This rejection is respectfully traversed.

The shortcomings of the Walsh disclosure are illustrated in Section 8, the Noda in Section 10, and the Genc disclosure in Section 5. Combination of the collected teachings fails to provide for a case of obviousness as Noda and Genc do not account for the flaws of the primary reference of Walsh. Namely, the reference combination lacks a passive vent, Walsh's water tank is not holding coolant – it is holding condensate, the Walsh water tank has the cathode gas stream ( $O_2$ ) fed into it, not the anode gas stream ( $H_2$ ), and the reference combination is silent with respect to using the collected water in the reservoir as a coolant.

As such, the present claims are not obvious over the combined references. Applicants request reconsideration of the claims and withdrawal of the rejection.

### **14. REJECTION UNDER 35 U.S.C. § 103 – WALSH, NODA, BUZZELLI & GENC**

Claim 18 stand rejected under 35 U.S.C. § 103(a) as allegedly unpatentable over Walsh

(U.S. Pub. No. 2003/0118881) in view of Noda (U.S. Pat. No. 5,623,390) and Buzzelli (U.S. Pat. No. 4,168,349) as applied to claim 17 and further in view of Genc (U.S. Pub. No. 2002/0160245). This rejection is respectfully traversed.

The shortcomings of the Walsh disclosure are illustrated in Section 8, the Noda disclosure in Section 10, the Buzzelli disclosure in Section 4, and the Genc disclosure in Section 5. Combination of the collected teachings fails to provide for a case of obviousness as Noda, Buzzelli, and Genc do not account for the flaws of the primary reference of Walsh. Namely, the reference combination lacks a passive vent, Walsh's water tank is not holding coolant – it is holding condensate, the Walsh water tank has the cathode gas stream ( $O_2$ ) fed into it, not the anode gas stream ( $H_2$ ), and the reference combination is silent with respect to using the collected water in the reservoir as a coolant.

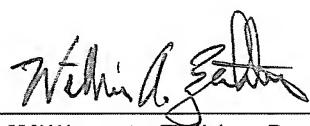
As such, the present claims are not obvious over the combined references. Applicants request reconsideration of the claims and withdrawal of the rejection.

## 15. CONCLUSION

It is believed that all of the stated grounds of rejection have been properly traversed, accommodated, or rendered moot. Applicant therefore respectfully requests that the Examiner reconsider and withdraw all presently outstanding rejections. It is believed that a full and complete response has been made to the outstanding Office Action and the present application is in condition for allowance. Thus, prompt and favorable consideration of this amendment is respectfully requested. If the Examiner believes that personal communication will expedite prosecution of this application, the Examiner is invited to telephone the undersigned at (248) 641-1600.

Respectfully submitted,

Dated: Aug. 14, 2009

By:   
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WAZ/akb

enclosure

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(12) **United States Patent**  
Wenderoth et al.

(10) **Patent No.:** US 7,393,464 B2  
(45) **Date of Patent:** Jul. 1, 2008

(54) **COOLING AGENTS FOR COOLING SYSTEMS IN FUEL CELL DRIVES**

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(\*) **Notice:** Subject to any disclaimer, the term of this patent is extended or adjusted under 35 U.S.C. 154(b) by 20 days.

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(22) **PCT Filed:** Dec. 14, 2001

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§ 371 (c)(1).

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PCT Pub. Date: Jul. 18, 2002

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(51) **Int. Cl.**

C09K 5/00 (2006.01)

H01M 8/18 (2006.01)

(52) **U.S. Cl.:** 252/71; 252/73; 252/75;  
429/20; 429/26; 429/120

(58) **Field of Classification Search:** 252/75,  
252/76, 78.3, 79, 387

See application file for complete search history.

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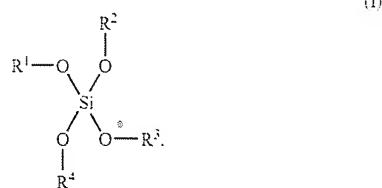
\* cited by examiner

Primary Examiner—Necholus Ogden

(74) **Attorney, Agent, or Firm:** Novak Druce + Quigg LLP

(57) **ABSTRACT**

The invention relates to antifreeze concentrates for cooling systems in fuel cell drives, from which are produced ready-to-use aqueous cooling agent compositions having a maximum conductivity of 50  $\mu$ s/cm, based on alkylene glycols or the derivatives thereof, and containing orthosilicic acid esters of formula (I) wherein the variables R<sup>1</sup> to R<sup>4</sup> are the same or different and represent C<sub>1</sub>-C<sub>20</sub> alkyl substituents, C<sub>2</sub>-C<sub>20</sub> alkylene substituents, C<sub>1</sub>-C<sub>20</sub> hydroxylalkyl substituents, optionally substituted C<sub>6</sub>-C<sub>12</sub> aryl substituents and/or glycol ether substituents of formula (CH<sub>2</sub>—CH<sub>2</sub>—O)<sub>n</sub>—R<sup>5</sup> wherein R<sup>5</sup> represents hydrogen or C<sub>1</sub>-C<sub>5</sub> alkyl and n represents a number between 1 and 5



14 Claims, No Drawings

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## COOLING AGENTS FOR COOLING SYSTEMS IN FUEL CELL DRIVES

The present invention relates to coolants for cooling systems in fuel cell drives, in particular for motor vehicles, based on alkylene glycols or derivatives thereof, which contain orthosilicic esters as corrosion inhibitors.

Fuel cells for mobile use in motor vehicles must be capable of being operated also at low outdoor temperatures down to about  $-40^{\circ}\text{C}$ .: a coolant circulation protected from freezing is therefore essential.

The use of radiator antifreezes conventionally used in internal combustion engines would not be possible in the case of fuel cells without complete electrical insulation of the cooling ducts, since, owing to the salts contained therein as corrosion inhibitors, these antifreezes have too high an electrical conductivity, which would adversely affect the operation of the fuel cell..

DE-A 198 02 490 (1) describes fuel cells having a cooling circulation which contains an antifreeze and in which a paraffinic isomer mixture having a pour point of less than  $-40^{\circ}\text{C}$ . is used as a coolant. However, the flammability of such a coolant is disadvantageous.

EP-A 1 009 050 (2) discloses a fuel cell system for automobiles, in which air is used as a cooling medium. The disadvantage here, however, is that air is known to be a poorer heat conductor than a liquid cooling medium.

WO 00/17951 (3) describes a cooling system for fuel cells, in which a pure monoethylene glycol/water mixture in the ratio 1:1, without additives, is used as a coolant. Since, owing to the absence of corrosion inhibitors, there has been no corrosion protection at all against the metals present in the cooling system, the cooling circulation contains an ion exchange unit to maintain the purity of the coolant and to ensure a low specific conductivity for a longer time, with the result that short-circuits and corrosion are prevented. Anionic resins, for example of the strongly alkaline hydroxyl type, and cationic resins, for example those based on sulfo groups, are mentioned as suitable ion exchangers, and other filtration units, for example active carbon filters, are mentioned.

The structure and the mode of operation of a fuel cell for automobiles, in particular a fuel cell comprising an electron-conducting electrolyte membrane (PEM fuel cell, polymer electrolyte membrane fuel cell) are described by way of example in (3), aluminum being a preferred metal component in the cooling circulation (radiator).

The use of silicon compounds, generally in the form of silicates, as corrosion inhibitors in radiator antifreezes for conventional internal combustion engines operated using gasoline or diesel fuel has long been known, for example from: G. Reinhard, "Aktiver Korrosionsschutz in wässrigen Medien", pages 87-98, expert-Verlag 1995 (ISBN 3-8169-1265-6).

EP-A 105 803 (4) discloses the use of orthosilicic esters in addition to ionic corrosion inhibitors in radiator antifreezes for automobiles having conventional gasoline or diesel internal combustion engines.

The use of orthosilicic esters as corrosion inhibitors in coolants for cooling systems in fuel cell drives is unknown to date.

A principal problem in the case of cooling systems in fuel cell drives is the maintenance of low electrical conductivity of the coolant in order to ensure safe and trouble-free operation of the fuel cell and permanently to prevent short-circuits and corrosion.

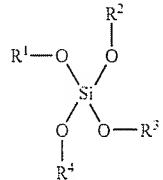
Surprisingly, it has now been found that the duration of low electrical conductivity in a cooling system based on alkylene

2

glycol/water, in particular if, according to (3), it contains an integrated ion exchanger, can be substantially increased by adding small amounts of orthosilicic esters: in practice, this has the advantage that the time intervals between two coolant changes in the case of fuel cell drives can be further extended, which is of interest particularly in the automotive sector.

Accordingly, we have found antifreeze concentrates for cooling systems in fuel cell drives, from which ready-to-use aqueous coolant compositions having a conductivity of not more than 50  $\mu\text{S}/\text{cm}$  result and which are based on alkylene glycols or derivatives thereof, which concentrates contain orthosilicic esters of the formula (I)

Formula (I)



where  $\text{R}^1$  to  $\text{R}^4$  are identical or different and are  $\text{C}_1$ - to  $\text{C}_{20}$ -alkyl,  $\text{C}_2$ - to  $\text{C}_{20}$ -alkenyl,  $\text{C}_1$ - to  $\text{C}_{20}$ -hydroxyalkyl, unsubstituted or substituted  $\text{C}_6$ - to  $\text{C}_{12}$ -aryl and/or a glycol ether substituent of the formula  $-(\text{CH}_2-\text{CH}_2-\text{O})_n-\text{R}^5$ , where  $\text{R}^5$  is hydrogen or  $\text{C}_1$ - to  $\text{C}_5$ -alkyl and  $n$  is from 1 to 5.

Preferred antifreeze concentrates here are those from which ready-to-use aqueous coolant compositions having a silicon content of from 2 to 2000, in particular from 10 to 1000, preferably from 25 to 500, especially from 40 to 250, ppm by weight, from orthosilicic esters of the formula I. result.

Typical examples of orthosilicic esters (I) used according to the invention are pure tetraalkoxysilanes, such as tetramethoxysilane, tetraethoxysilane, tetra-n-propoxysilane, tetraisopropoxysilane, tetra-n-butoxysilane, tetra-tert-butoxysilane, tetra(2-ethylbutoxy)silane and tetra(2-ethylhexyloxy)silane, and furthermore tetraphenoxyxilane, tetra(2-methylphenoxy)silane, tetravinylxilane, tetraallyloxyxilane, tetra(2-hydroxyethoxy)silane, tetra(2-ethoxyethoxy)silane, tetra(2-butoxyethoxy)silane, tetra(1-methoxy-2-propoxy)silane, tetra(2-methoxyethoxy)silane and tetra[2-[2-(2-methoxyethoxy)ethoxy]ethoxy]silane. The orthosilicic esters (I) used preferably have four identical variables  $\text{R}^1$  to  $\text{R}^4$ .

In a preferred embodiment, orthosilicic esters (I) in which  $\text{R}^1$  to  $\text{R}^4$  are identical and are  $\text{C}_1$ - to  $\text{C}_4$ -alkyl or a glycol ether substituent of the formula  $-(\text{CH}_2-\text{CH}_2-\text{O})_n-\text{R}^5$ , where  $\text{R}^5$  is hydrogen, methyl or ethyl and  $n$  is 1, 2 or 3, are used.

Said orthosilicic esters (I) are either commercially available or can be prepared by simple transesterification of one equivalent of tetramethoxysilane with four equivalents of the corresponding longer-chain alcohol or phenol and by distilling off methanol.

Ready-to-use aqueous coolant compositions which have a conductivity of not more than 50  $\mu\text{S}/\text{cm}$  and substantially comprise

- (a) from 10 to 90% by weight of alkylene glycols or derivatives thereof,
- (b) from 90 to 10% by weight of water and

(c) from 2 to 2000, preferably from 25 to 500, ppm by weight of silicon from orthosilicic esters of the formula I

can be prepared from the antifreeze concentrates by dilution with ion-free water. The sum of all components is 100% by weight here.

The present invention thus also relates to ready-to-use aqueous coolant compositions for cooling systems in fuel cell drives, which substantially comprise

(a) from 10 to 90% by weight of alkylene glycols or derivatives thereof.

(b) from 90 to 10% by weight of water and

(c) from 2 to 2000, preferably from 25 to 500, ppm by weight of silicon from orthosilicic esters of the formula I

and which are obtainable by diluting said antifreeze concentrates with ion-free water. The sum of all components is 100% by weight here.

The novel ready-to-use aqueous coolant compositions have an initial electrical conductivity of not more than 50, in particular 25, preferably 10, especially 5,  $\mu$ S/cm. The conductivity is kept at this low level in continuous operation of the fuel cell drive over several weeks or months, particularly if a cooling system comprising integrated ion exchanger is used in the fuel cell drive.

The pH of the novel ready-to-use aqueous coolant compositions decreases over the operating time substantially more slowly than in the case of cooling liquids not containing added orthosilicic esters. The pH is usually from 4.5 to 7 in the case of fresh coolant compositions according to the invention and can decrease to 3.5 in continuous operation.

The ion-free water used for dilution may be pure distilled or bidistilled water or, for example, water demineralized by ion exchange.

The preferred weight ratio in which an alkylene glycol or a derivative thereof is mixed with water in the ready-to-use aqueous coolant compositions is from 25:75 to 80:20, in particular from 35:65 to 75:25, preferably from 50:50 to 70:30, especially from 55:45 to 65:35. In particular, monoethylene glycol, but also monopropylene glycol, polyglycols, glycol ethers or glycerol, in each case alone or as mixtures thereof, can be used as alkylene glycol components or derivatives thereof. Monoethylene glycol alone or mixtures containing monoethylene glycol as the main component, i.e. having a content of more than 50, in particular more than 80, especially more than 95, % by weight in the mixture, with other alkylene glycols or derivatives of alkylene glycols are particularly preferred.

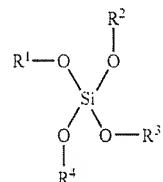
The dosage of the respective orthosilicic esters (I) in the ready-to-use aqueous coolant compositions is calculated from the above data by means of the silicon content based on (I).

The novel antifreeze concentrates themselves, from which the described ready-to-use aqueous coolant compositions result, can be prepared by dissolving the orthosilicic esters (I) in alkylene glycols or derivatives thereof, which may be used

in anhydrous form or with a low water content (up to about 10, in particular up to 5, % by weight).

The present invention also relates to the use of orthosilicic esters of the formula I

Formula (I)



where R<sup>1</sup> to R<sup>4</sup> are identical or different and are C<sub>1</sub>- to C<sub>20</sub>-alkyl, C<sub>2</sub>- to C<sub>20</sub>-alkenyl, C<sub>1</sub>- to C<sub>20</sub>-hydroxyalkyl, unsubstituted or substituted C<sub>6</sub>- to C<sub>12</sub>-aryl and/or a glycol ether substituent of the formula —(CH<sub>2</sub>—CH<sub>2</sub>—O)<sub>n</sub>—R<sup>5</sup>, where R<sup>5</sup> is hydrogen or C<sub>1</sub>- to C<sub>5</sub>-alkyl and n is from 1 to 5.

for the preparation of antifreeze concentrates for cooling systems in fuel cell drives, in particular for motor vehicles, based on alkylene glycols or derivatives thereof.

The present invention furthermore relates to the use of the antifreeze concentrates described for the preparation of ready-to-use aqueous coolant compositions having a conductivity of not more than 50  $\mu$ S/cm for cooling systems in fuel cell drives, in particular for motor vehicles.

## EXAMPLES

The examples which follow illustrate the invention without restricting it.

The novel coolant compositions were tested with regard to their suitability for fuel cell drives by the test described below, in comparison with a coolant composition according to (3):

### Description of Test:

Five aluminum test metals (vacuum-soldered Al, designation: EN-AW 3005, solder-plated on one side with 10% by weight of EN-AW 4045; dimensions: 58×26×0.35 mm, having a hole of 7 mm diameter) were weighed, connected non-conductively by means of a plastics screw with nut and Teflon washer and placed on two Teflon supports in a 1 l beaker with ground glass joint and glass cover. Thereafter, 1000 ml of test liquid were introduced and a small fabric bag containing 2.5 g of an ion exchanger (mixed bed ion exchanger resin AMBERJET UP 6040 RESIN from Rohm+Haas) was suspended in the liquid. The beaker was closed air-tight with the glass cover and heated to 88° C. and the liquid was stirred vigorously with a magnetic stirrer. The electrical conductivity was measured at the beginning of the test and after 7 and 42 (or after 77) days (conductivity meter LF 530 from WTW/Weilheim). The test was then terminated: the aluminum samples were assessed visually and, after pickling with aqueous chromic acid/phosphoric acid, were evaluated gravimetrically according to ASTM D 1384-94.

The results are shown in Table 1 below.

TABLE

Coolant composition:	Comparative example (according to WO 00/17951):	Example 1:	Example 2:
	60% by volume of monoethylene glycol 40% by volume of water	60% by volume of monoethylene glycol 40% by volume of water 742 ppm by weight of tetraethoxy-silane	60% by volume of monoethylene glycol 40% by volume of water 3600 ppm by weight of tetra[2-[methoxyethoxy]ethoxy]ethoxy-silane
Electrical conductivity [ $\mu\text{S}/\text{cm}$ ]			
Beginning of test:	2.0	0.8	2.6
After 7 days:	2.3	0.8	2.2
After 42 days:	36.2	3.0	14.4
After 77 days:	—	—	18.6
pH:			
Beginning of test	6.9	6.6	4.7
End of test:	2.9	4.0	3.6
Appearance	slightly stained	stained	stained
Aluminum samples after the test:			
Weight change [ $\mu\text{g}/\text{cm}^2$ ]			
After pickling:			
1	-0.05	-0.02	-0.02
2	-0.04	-0.01	-0.02
3	-0.04	-0.02	-0.04
4	-0.04	-0.02	-0.04
5	-0.03	-0.02	-0.04
Mean value of the 5 samples	-0.04	-0.02	-0.03
Solution after end of test	yellowish, clear	colorless, clear	yellowish, clear

In the mixture of monoethylene glycol and water, the volume ratio of 60:40 corresponds to a weight ratio of 62.5:37.5.

In the novel examples 1 and 2, the orthosilicic esters were metered so that a silicon content of 100 ppm by weight in each case was present in the cooling liquid.

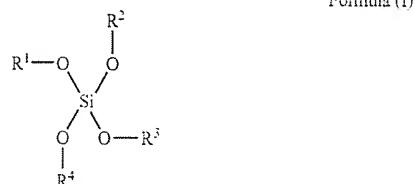
The results show that a very low electrical conductivity of less than 5  $\mu\text{S}/\text{cm}$  was still present even after an uninterrupted test period of 42 days in the case of the novel example 1, whereas a substantial deterioration had occurred, with an increase to virtually 40  $\mu\text{S}/\text{m}$ . in the case of the additive-free coolant according to WO 00/17951 (3). However, with the novel example 2 which was slightly poorer compared with example 1 after 42 days, the specific conductivity was still about 50% lower even after a test period of 72 days than in the case of the comparative example after a test duration of 42 days.

In no case did significant corrosion occur on the aluminum samples.

What is claimed is:

1. A method of protecting fuel cell drives from corrosion using cooling systems which are based on antifreeze concentrates based on alkylene glycols or derivatives thereof, from which ready-to-use aqueous coolant compositions having a

conductivity of not more than 50  $\mu\text{S}/\text{cm}$  result, wherein the anti-freeze concentrates contain orthosilicic esters of the formula I



15 where  $\text{R}^1$  to  $\text{R}^4$  are identical or different and are  $\text{C}_1$ - to  $\text{C}_{20}$ -alkyl,  $\text{C}_2$ - to  $\text{C}_{20}$ -alkenyl,  $\text{C}_1$ - to  $\text{C}_{20}$ -hydroxyalkyl, unsubstituted or substituted  $\text{C}_6$ - to  $\text{C}_{12}$ -aryl and/or a glycol ether substituent of the formula  $-(\text{CH}_2-\text{CH}_2-\text{O})_n-\text{R}^5$ , where  $\text{R}^5$  is hydrogen or  $\text{C}_1$ - to  $\text{C}_5$ -alkyl and  $n$  is from 1 to 5.

20 2. A method as claimed in claim 1, wherein the antifreeze concentrates result in ready-to-use aqueous coolant compositions having a silicon content of from 2 to 2000 ppm by weight, from orthosilicic esters of the formula I.

25 3. A method as claimed in claim 1, wherein the antifreeze concentrates contain orthosilicic esters of the formula I, in which  $\text{R}^1$  to  $\text{R}^4$  are identical and are  $\text{C}_1$ - to  $\text{C}_2$ -alkyl or a glycol ether substituent of the formula  $-(\text{CH}_2-\text{CH}_2-\text{O})_n-\text{R}^5$ , where  $\text{R}^5$  is hydrogen, methyl or ethyl and  $n$  is 1, 2 or 3.

30 4. A method as claimed in claim 1, wherein the alkylene glycol used for the antifreeze concentrates is monoethylene glycol.

35 5. A method as claimed in claim 1, wherein ready-to-use aqueous coolant compositions which have a conductivity of not more than 50  $\mu\text{S}/\text{cm}$  and substantially comprise

(a) from 10 to 90% by weight of alkylene glycols or derivatives thereof,

(b) from 90 to 10% by weight of water and

(c) from 2 to 2000 ppm by weight of silicon from orthosilicic esters of the formula I

40 are produced from the antifreeze concentrates by dilution with ion-free water.

45 6. The method of claim 2 wherein the silicon content is from 25 to 500 ppm.

7. The method of claim 5 wherein the silicon content is from 25 to 500 ppm.

45 8. The method of claim 1, wherein the cooling system is in a fuel cell which contains an electron-conducting electrolyte membrane.

50 9. The method of claim 1, wherein the conductivity of the ready-to-use aqueous coolant compositions is not more than 25  $\mu\text{S}/\text{cm}$ .

10. The method of claim 1, wherein the conductivity of the ready-to-use aqueous coolant compositions is not more than 10  $\mu\text{S}/\text{cm}$ .

55 11. The method of claim 1, wherein the conductivity of the ready-to-use aqueous coolant compositions is not more than 5  $\mu\text{S}/\text{cm}$ .

12. The method of claim 5, wherein the conductivity of the ready-to-use aqueous coolant compositions is not more than 25  $\mu\text{S}/\text{cm}$ .

13. The method of claim 5, wherein the conductivity of the ready-to-use aqueous coolant compositions is not more than 10  $\mu\text{S}/\text{cm}$ .

60 14. The method of claim 5, wherein the conductivity of the ready-to-use aqueous coolant compositions is not more than 5  $\mu\text{S}/\text{cm}$ .